This Page Is Inserted by IFW Operations and is not a part of the Official Record

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

IMAGES ARE BEST AVAILABLE COPY.

As rescanning documents will not correct images, please do not report the images to the Image Problem Mailbox.



PCT

WORLD INTELLECTUAL PROPERTY ORGANIZATION International Bureau

INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 6:
H01B 3/44

A1

(11) International Publication Number: WO 96/37899

(43) International Publication Date: 28 November 1996 (28.11.96)

(21) International Application Number: PCT/GB96/01252 (81) Désignated States: AL, AM, AT, AU, AZ, BB, BG, BR, BY,

21) Michalional Application Manuel.

(22) International Filing Date: 24 May 1996 (24.05.96)

9510620.9 25 May 1995 (25.05.95) GB

(71) Applicant (for all designated States except US): BICC PUBLIC LIMITED COMPANY [GB/GB]; Devonshire House, Mayfair Place, London W1X 5FH (GB).

(71) Applicant (for US only): KATZ, Maurice (legal representative of the deceased inventor) [GB/AU]; 2 Rosslyn Court, Seaford, VIC 3198 (AU).

(72) Inventor: PANASIUK, Leon (deceased).

(72) Inventors; and

(30) Priority Data:

(75) Inventors/Applicants (for US only): VERNE, Stefan [GB/GB]; 3 Brunswick Gardens, London W5 1AP (GB). BIELSTEIN, George, Paul, Richard [GB/GB]; 14 Temple Sheen, London SW14 7RP (GB).

(74) Agent: POOLE, Michael, John; BICC Patents & Licensing Dept., Quantum House, Maylands Avenue, Hemel Hempstead, Hertfordshire HP2 4SJ (GB).

(81) Designated States: AL, AM, AT, AU, AZ, BB, BG, BR, BY, CA, CH, CN, CZ, DE, DK, EE, ES, FI, GB, GE, HU, IS, IP, KE, KG, KP, KR, KZ, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, TJ, TM, TR, TT, UA, UG, US, UZ, VN, ARIPO patent (KE, LS, MW, SD, SZ, UG), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).

Published

With international search report.

(54) Title: POLYMER COMPOSITIONS AND ELECTRIC CABLES

(57) Abstract

An environmentally desirable composition with sufficient electrical stability to be suitable for making the dielectric (insulation) of a medium-voltage cable comprises, in parts by weight polymer base, of which at least 50 parts are ethylene copolymer rubber and the balance, if any, polyethylene 100, mineral filler of which at least half is calcined clay 40-120, surface treatment agent for the filler 1-6, zinc oxide 10-30, lead compounds not to exceed 1 calculated as element and appropriate amounts of conventional ingredients comprising an antioxidant, a curing system and a processing aid.

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AM	Armenia	GB	United Kingdom	MW	Malawi
AT	Austria	GE	Georgia	MX	Mexico
ΑU	Australia	GN	Guinea	NE	Niger
BB	Barbados	GR	Greece	NL	Netherlands
BE	Belgium	HU	Hungary	NO	Norway
BF	Burkina Faso	IE	Ireland	NZ	New Zealand
BG	Bulgaria	IT	Italy	PL	Poland
BJ	Benin	, JP	Japan	PT	Portugal
BR	Brazil	KE	Kenya	RO	Romania
BY	Belarus	KG	Kyrgystan	RU	Russian Federation
CA	Canada	KP	Democratic People's Republic	\$D	Sudan
CF	Central African Republic		of Korea	SE	Sweden
CG	Congo	KR	Republic of Korea	SG	Singapore
CH	Switzerland	KZ	Kazakhstan	SI	Slovenia
CI	Côte d'Ivoire	LI	Liechtenstein	SK	Slovakia
CM	Cameroon	LK	Sri Lanka	SN	Senegal
CN	China	LR	Liberia	SZ	Swaziland
CS	Czechoslovakia	LT	Lithuania	TD	Chad
cz	Czech Republic	LU	Luxembourg	TG	Togo
DE	Germany	LV	Latvia	TJ	Tajikistan
DK	Denmark	MC	Monaco	TT	Trinidad and Tobago
EE	Estonia	MD	Republic of Moldova	UA	Ukraine
ES	Spain ·	MG	Madagascar	UG ·	Uganda
FI	Finland	ML	Mali	US	United States of America
FR	Prance	MN	Mongolia	UZ	Uzbekistan
GA.	Gabon	MR	Mauritania	VN	Viet Nam

Polymer Compositions and Electric Cables

This invention relates to polymer compositions for use in electric cables, and more particularly as dielectric (insulation) in "medium voltage" power cables in which the dielectric is bounded by a conductor screen and a core screen both of conductive polymeric material; it also relates to the cables in which they are used.

More particularly, it relates to compositions based on ethylene copolymer rubbers, which may be defined for 10 certainty in the context of this application as hydrocarbon copolymers of ethylene with at least one other alkene monomer having from 3 to 10 carbon atoms which copolymers have rubbery elastic characteristics.

Until recently, only two ethylene copolymer rubbers

15 have been widely used in such compositions, namely EPR, in
which the major repeating units are ethylene and propylene,
and EPDM, in which a minor proportion of a diene monomer
(such as norbornadiene) is present in addition to ethylene
and propylene. More recently, the development of "single
20 site" catalysts has made available additional ethylene
copolymer rubbers which appear to be based predominantly on
ethene and octene or on ethylene and hexene.

The service life of medium-voltage cables is in many cases limited by the long term diminution of electric

25 strength of the dielectric which is now attributed to the formation of water-trees (or electric trees, if water is absent), and current EPR formulations generally contain an oxide of lead in an amount corresponding to several phr (parts per hundred of polymer) of lead, calculated as

30 element, as the means of securing a sufficient stability of electrical strength (a tree suppressant). Although the amount of lead used in electric cables is small compared with other usages, it is now considered desirable to eliminate any

2

avoidable source of lead in contact with the environment.

Zinc oxide is a conventional constituent of EPR- and EPDM-based compositions, used at a level around 5phr as a curing aid or pH stabiliser; but it has not been perceived as 5 having a useful effect on electrical stablility.

Partly as a result of a review of unpublished experiments carried out in our laboratories by the late Leon Panasiuk under the supervision of Dr George P R Bielstein and Mr Stefan Verne between about 1964 and 1967, we have now recognised that higher levels of zinc oxide lead to useful levels of electrical stability under wet conditions, even in compositions in which the content of lead compounds is below 1%.

The compositions in accordance with the present invention comprise, in parts by weight:

Polymer base, of which at least 50 parts are ethylene copolymer rubber and the balance, if any,

polyethylene 100

20 Mineral filler of which at least

half is calcined clay 40-120

Surface treatment agent for

the filler 1-6

Zinc oxide 10-30

25 Lead compounds not to exceed 1 calculated as element

and appropriate amounts of conventional ingredients comprising an antioxidant, a curing system and a processing aid.

The ethylene copolymer rubber may be an EPR or an EPDM, each of which may be either semi-crystalline or

3

amorphous provided that it has rubbery elastic properties. Ethylene copolymer rubbers made with "single-site" catalyst systems can also be used and may have advantages in some cases.

The surface treatment agent for the filler is preferably selected from silanes, siloxanes, metal salts of organic acids, and organic acids that are substantially insoluble in water. More than one surface treatment agent can be used. Silane surface treatment agents are preferred. The filler is preferably pre-treated with the surface treatment agent (or with one of them) but preferably the composition includes free surface treatment agent as well. Appropriate amounts will vary somewhat from one surface treatment agent to another, but will typically be in the range from about 1-2 parts pre-coated on the filler and about 1-5 parts added separately.

Preferred content of zinc oxide is about 15-25 parts; conventional grades can be used, but grades of higher purity 20 are preferred.

Compositions based on semicrystalline EPR's or EPDM's preferably contain polyethylene, preferably in an amount in the range from 3 to 10 parts, as a processing aid; larger amounts, up to the limit of 50 parts, can be used for cost reduction and/or in the case of some amorphous EPR's or EPDM's to enable granulation of the composition.

From an environmental viewpoint, the content of lead compounds should be as small as possible, and ideally zero; however, a small amount of certain lead compounds may achieve a major improvement in electrical stability with much less environmental hazard than conventional EPR or EPDM

compositions for medium-voltage cables. In particular, there may be merit in using up to about 2 parts of lead stearate (that is up to about 0.54 parts calculated as element).

Any of the conventional antioxidants can be used in 5 their usual amounts; we particularly prefer antioxidants of the poly-dihydroxyquinoline class, in amounts in the range 0.5 to 3 parts.

Conventional curing systems can be used; in particular, all the compositions of the invention can be cured with peroxides, optionally with poly-unsaturated co-agents such as triallylisocyanurate, or with peroxides and sulfur; compositions based on EPDM's can also be cured with conventional sulfur-based systems. No change in curing practice is expected to be needed.

Suitable processing aids, apart from those already mentioned, include paraffin wax in conventional amounts up to around 6 parts and hydrocarbon oils in amounts up to around 30 parts (varying form oil to oil).

EXAMPLES

Compositions as detailed below were prepared on a 12-inch (300m) laboratory 2-roll mill; the polymer-base was plasticised and the other ingredients, except curing agents, added at a temperature controlled to be in the range 120-130°; the curing agents were added in a separate step on the "cold" mill. It should be noted that a few ingredients used in experiments around 30 years ago are no longer available. Example 1

To 95 parts of an EPDM (Royalene® 201) and 5 parts of a polyethylene(WJG11) were added: 67 parts calcined clay

30 (Whitetex®) and 48 parts talc (Mistron Vapour®) as additional mineral filler; 1 part vinyl tris(2-methoxyethoxy) silane

5

(Silane A172®) as surface treatment agent; 14 parts zinc oxide; ; 1 part of polymerised trimethyldihydroquinoline (Flectol H®) and 1.3 parts of N-nitroso-N-methyl-p-nitrosoaniline as antioxidants; a curing system comprising 1 part dicumyl peroxide and 1 part N,N'(1,3-phenylene) dimaleimide; and 9.5 parts of Esso® Oil D and 5 parts of paraffin wax as processing aids. After press cure for 45 minutes at 160°, the composition had a tensile strength of 8.48MPa (1230 psi), elongation (meaning elongation at break) 10 of 250% and volume resistivity of 4.9 x 10¹⁵ ohm.cm.

In preliminary ageing tests in air at 150°C, the composition showed first signs of brittleness after about 35 days.

Electrical aging tests were also carried out using the 15 material extruded to a minimum radial thickness of 0.76mm (0.030 inch) onto a strand comprising three tinned copper wires each 0.74mm (0.029inch) in diameter. The insulated strand was immersed in water at 75°C and exposed continuously (except during measurements) to a voltage of 600V at 50Hz; at 20 intervals, insulation resistance was measured at 500V dc one minute after application of this voltage and specific inductive capacitance and power factor at 1.2kV 50Hz. Within one day's exposure, the volume resistivity had fallen to 1.5 x 10¹⁴ ohm.cm, but this then remained substantially 25 unchanged (readings ranged from 1.0 to 2.6 \times 10¹⁴) up to and including the 59th week. Deterioration was detectable in about weeks 60 to 70, when readings ranged from 0.49 to 1.0 x 10¹⁴ ohm.cm, and gradually increased with the readings falling below 0.1×10^{14} ohm.cm for the first time at week 74 30 and reaching 0.014 x 10¹⁴ when the tests were discontinued at

120 weeks. Specific inductive capacitance varied by no more than 5% from its value after one day's exposure until week 78, and had changed by only about 8% at the end of 120 weeks. Power factor changed little during the tests; after 5 one week, it was in the range 6-7.2%, where it remained for 19 weeks; it then slightly reduced, falling below 3% at week 32 and remaining between 2.0 and 3.0 until week 57, when a slow rise was observed, reaching 4% at week 94, 4.8 at week 112 and then rising more rapidly to 5.3 at week 114, 6.8 at week 118 and 7.7 at the end of the test at 120 weeks.

Example 2

This formulation was similar to Example 1 but 1 part of oleic acid was added as an additional surface treatment agent for the filler. After press cure for 45 minutes at 160°, the

- 15 composition had a tensile strength of 7.07MPa (1025psi), elongation of 390% and volume resistivity of 13 x 10¹⁵ ohm.cm. This composition showed first signs of brittleness in the preliminary air ageing test after 48 days at 150°C. The volume resistivity in the wet electrical
- ageing test remained in the range 0.9 to 2.3 x 10^{14} ohm.cm from about 3 to 43 weeks and then slowly declined, reaching 0.01 x 10^{14} ohm.cm at about 52 weeks and 0.007 x 10^{14} ohm.cm when the test was discontinued at 71 weeks. Specific inductive capacitance changed by 5.2% within 1 week and then
- 25 increased slowly and fairly steadily to have changed about 16% after 59 weeks; lower change values (in the range 5-10%) were obtained in weeks 61-71, but may be suspect.

Example 3

To 100 parts of an experimental grade of EPDM (DuPont® ECD-30 618-60) were added 120 parts of a proprietory silanepretreated calcined clay (Translink® 37), 1 part of stearic acid as an additional surface treatment agent for the clay, 20 parts of zinc oxide, 1 part of a primary aromatic amine antioxidant (LD-382-30), a curing system comprising 1.25 parts sulfur, 3 parts of a proprietory mixture (EPTAC.2) comprising equal weights of zinc dimethyl dithiocarbamate and tetra-methyl thiuram disulfide, 2 parts of 2-mercaptobenzothiazole and 1.3 parts of 75%-active tellurium diethyl dithocarbamate(PTD-75) and as processing aids 30 parts of Sunpar® oil 2280 and 3 parts paraffin wax. After press cure for 45 minutes at 160°, the composition had a tensile strength of 4.4MPa (645psi) and an elongation of 430%. This composition did not show signs of brittleness in the air ageing test until the 60th day.

The composition was made into an insulated wire sample and tested as before, and the insulation resistance observed remained in the range 2.6 to 4.9 x 10¹⁴ ohm.cm throughout 39 weeks of observation; the specific inductive capacity changed gradually over about 21 weeks and then remained fairly stable with the change remaining in the range 7.6 to 10.2% for the remainder of the test; power factor was essentially constant in the range 0.9 to 1.2% throughout the test.

Example 4

This was identical with Example 1, except that 1 part of lead 25 stearate was added. After press cure for 45 minutes at 160°, the composition had a tensile strength of 6.4MPa (925psi), elongation of 515% and volume resistivity of 9.2 x 10¹⁵ ohm.cm.

In this case, the volume resistivity remained in the 30 range 1.1 to 3.6×10^{14} ohm.cm throughout the 120-week period

8

of the test in water, with values in the upper part of the range (over 2.5) observed mostly in the period from 34 to 68 weeks. Measurements of increase in specific inductive capacitance showed no very clear pattern, but were mostly 5 below 5% for the first 64 weeks and mostly in the range 5-10% in weeks 65-120. Power factors observed were all in the range from 1.7 to 2.8, with only about five readings over

Example 5

2.0% after week 30.

- 10 The composition of this example comprises 95 parts of a semicrystalline EPR (Nordel® 2722), 5 parts of a low-density polyethylene of Melt Index 2.0, 60 parts of surface-treated calcined clay (Translink® 37), 1 part of vinyl tris(2-methoxy ethoxy) silane, 20 parts zinc oxide, 5 parts paraffin wax,
- 15 1.5 parts antioxidant (Flectol® H) and 2.5 parts of dicumyl peroxide (Dicup®).

Example 6

The composition of this example comprises 100 parts of an amorphous EPR (Nordel® 2522), 60 parts of surface-treated 20 calcined clay (Translink® 37), 1 part of vinyl tris(2-methoxy ethoxy) silane, 20 parts zinc oxide, 5 parts paraffin wax, 1.5 parts antioxidant (Flectol® H) and 2.5 parts of dicumyl peroxide (Dicup®).

Example 7

25 The composition of this example comprises 100 parts of an semicrystalline EPR (Nordel® 2722), 60 parts of surface-treated calcined clay (Translink® 37), 1 part of vinyl tris(2-methoxy ethoxy) silane, 20 parts zinc oxide, 1.5 parts paraffin wax, 1.5 parts antioxidant (Agerite® MA) and 2.5

30 parts of dicumyl peroxide (Dicup®).

9

Example 8

This was identical with Example 7 except that the clay was a non-commercial untreated one, identical with Translink 37 except for the omission of the surface treatment normally 5 applied to it.

Example 9

This was similar to Example 7 except that 40 parts of a polyethylene with Melt Index 2.0 and density about 0.92 was substituted for an equal number of parts of the EPR and that 10 the content of zinc oxide was only 10 parts.

Comparison Examples A and B

These formulations were similar to Example 1 except that the content of zinc oxide was only 5 parts and that there were minor differences in the antioxidant and curing systems: in both cases, half of the Flectol® H was replaced with an equal weight of a third antioxidant, zinc-2-mercaptobenzimidazolate; the curing systems comprised:

Comparison Example A sulfur (1 part);
tellurium diethyl dithiocarbamate

(Tellurac®) (1.5 parts, 80% active);
dipentamethylene thiuram
tetrasulfide (Tetrone® A)
(1.5 parts); and
zinc dimethyl dithiocarbamate

(0.5 part) -

Comparison Example B dicumyl peroxide (Dicup®) (3 parts) and sulfur (0.3 part).

Both these comparison examples were rejected on the basis of preliminary screening tests in air at 150°C, and were not subjected to electrical stability tests; comparison Example A exhibited brittleness in this test at 11 days and Comparison

10

Example B surface degradation at 6 days.

Comparison Example C

This was identical with Example 1 except that a conventional 3 parts of litharge was added. The composition showed 5 embrittlement in the air ageing test at 48 days (rather better than Example 1, but no better than wholly lead-free Example 2). In the electrical stability test, the volume

resistivity slowly rose from an intial value of about 1.2×10^{14} ohm.cm to reach 3 $\times 10^{14}$ ohm.cm about week 36,

10 remaining fairly stable to week 60 and then declining steadily to about 0.03 x 10¹⁴ ohm.cm at the end of 120 weeks. Specific inductive capacitance changes for this composition were mearly all negative, ranging from +0.4 (at one week) to -7.5 (at 66 weeks) without any clear pattern. Power factor

15 was similary almost constant, ranging from 1.1 to 4.7.

Comparison Example D

This is similar to Example 3 except for the addition of 4 parts of lead dioxide and the use of a different curing system consisting of 1 part dicumyl peroxide and 1 part 20 N,N'-(1,3 phenylene) dimaleimide. This composition showed signs of surface degradation in the air aging test after 34 days. In the electrical stability test, the volume resistivity was higher than for Example 3, ranging from 4.4 to 6.7 x 10¹⁴ ohm.cm over weeks 1-39, but there was no noticeable difference in stability; changes in specific inductive capacitance were also smaller (not over 4.3%) with the highest values (over 2.0%) observed in weeks 6 to 33. Power factor was substantially constant and a little better than for Example 3, with values from 0.4 to 0.6% recorded.

WO 96/37899

11

CLAIMS

A composition suitable for making the dielectric of a medium-voltage electric cable comprising, in parts by weight:

Polymer base, of which at least

5 50 parts are ethylene copolymer rubber and the balance, if any, polyethylene

100

PCT/GB96/01252

Mineral filler of which at least

half is calcined clay

40-120

10 Surface treatment agent for

the filler 1-6
Zinc oxide 10-30

Lead compounds not to exceed 1 calculated as element

- 15 and appropriate amounts of conventional ingredients comprising an antioxidant, a curing system and a processing aid.
 - 2 A composition as claimed in claim 1 in which the ethylene copolymer rubber is an EPR or an EPDM.
- 20 3 A composition as claimed in claim 1 in which the ethylene copolymer rubber is one made with "single-site" catalyst system.
 - 4 A composition as claimed in any one of claims 1-3 in which all of the filler is calcined clay.
- 25 A composition as claimed in any one of claims 1-4 in which the surface treatment agent for the filler is selected from silanes, siloxanes, metal salts of organic acids, and organic acids that are substantially insoluble in water.
- 6 A composition as claimed in any one of claims 1-5 in 30 which the filler is pre-treated with the surface treatment agent.

12

- A composition as claimed in claim 6 which also includes free surface treatment agent.
- 8 A composition as claimed in any one of claims 1-7 in which the content of zinc oxide is about 15-25 parts.
- 5 9 A composition in accordance with any one of claims 1-8 based on a semicrystalline EPR or EPDM and containing 3 to 10 parts polyethylene.
 - 10 A lead-free composition in accordance with any one of claims 1-9.
- 10 11 A composition in accordance with any one of claims 1-9 comprising up to about 2 parts of lead stearate.
 - 12 A medium-voltage electric cable having dielectric made from the composition claimed in any one of claims 1-11.
 - 13 A lead-free composition suitable for making the
- 15 dielectric of medium-voltage cable substantially as described with reference to either Example 5 or Example 6.
 - 14 A lead-free composition suitable for making the dielectric of medium-voltage cable substantially as described with reference to any one of Examples 7-9.

INTERNATIONAL SEARCH REPORT

Inter Total Application No PC1/GB 96/01252

A. CLASSII IPC 6	FICATION OF SUBJECT MATTER H01B3/44				
A	International Patent Classification (IPC) or to both national class	fication and IPC			
		nessed and if C			
	SEARCHED ocumentation searched (classification system followed by classification)	tion symbols)			
IPC 6	H01B				
Documentati	ion searched other than minimum documentation to the extent that	such documents are included in the fields s	arched		
Electronic d	ata base consulted during the international search (name of data ba	se and, where practical, search terms issed)			
C. DOCUM	MENTS CONSIDERED TO BE RELEVANT				
Category *	Citation of document, with indication, where appropriate, of the	relevant passages	Relevant to claim No.		
X	EP 0 042 440 A (DAINICHI NIPPON LTD) 30 December 1981 see page 20, line 11 - line 17 additive 29 see page 21 see page 23 - page 24; examples see page 37; example 38 see claims 1-19		1-14		
A	FR 2 344 613 A (ALCAN RES & DEV) October 1977 see claims 1-18; examples 15-19 US 4 204 024 A (PYE RICHARD T) 2 see claims 1-12		1-14		
Fu	of the documents are listed in the continuation of box C.	X Patent family members are listed	in annex.		
	- Carlot de Carl				
'A' document defining the general state of the art which is not considered to be of particular relevance		or priority date and not in conflict or cited to understand the principle or invention	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention (** document of particular relevance; the claimed invention		
filing	r document but published on or after the international g date	cannot be considered novel or cann	ot be considered to		
whic	ment which may throw doubts on priority claim(s) or h is cited to establish the publication date of another	involve an inventive step when the document is taken alone 'document of particular relevance; the daimed invention cannot be considered to involve an inventive step when the			
O, qoor	ion or other special reason (as specified) ment referring to an oral disclosure, use, exhibition or	document is combined with one or ments, such combination being obv	more other such docu-		
P' docu	r means ment published prior to the international filing date but than the priority date claimed	in the art. *&* document member of the same pate			
	ne actual completion of the international search	Date of mailing of the international	search report 3. 09. 96		
	22 August 1996	•	J. UJ. JU		
Name and	d mailing address of the ISA	Authorized officer			
	European Palent Office, P.B. 5818 Patentiaan 2 NL - 2280 HV Rijswijk Tel. (+ 31-70) 340-2040, Tx. 31 651 epo nl, Fax (+ 31-70) 340-3016	Stienon, P			

INTERNATIONAL SEARCH REPORT

aformation on patent family members

Inter 'onal Application No PC I/GB 96/01252

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP-A-0042440	30-12-81	JP-C- 1454497 JP-A- 56090845 JP-B- 63001340 AU-B- 6645181 CH-A- 651312 DE-C- 3050176 DE-T- 3050176 GB-A,B 2080314 WO-A- 8101852 SE-B- 447905 SE-A- 8105038 US-A- 4410648	25-08-88 23-07-81 12-01-88 22-07-81 13-09-85 15-11-90 25-03-82 03-02-82 09-07-81 22-12-86 25-08-81 18-10-83
FR-A-2344613	14-10-77	CA-A- 1095677 DE-A- 2711260 GB-A- 1565402 GB-A- 1565403 JP-C- 1518073 JP-A- 52115851 JP-B- 63060062 NL-A- 7702764 US-A- 4161419	17-02-81 22-09-77 23-04-80 23-04-80 07-09-89 28-09-77 22-11-88 19-09-77 17-07-79
US-A-4204024	20-05-80	NONE	